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FINAL TECHNICAL REPORT

for the grant titled:

THE PHYSICS OF SPIN POLARIZED GASES

Grant AFOSR-F49620-92-J-0211

Research Period:
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Submitted January 16, 1995
by
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Note: W. Happer is the principal investigator to the follow-on to this grant.

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I. Introduction

Most of our research was connected either directly or indirectly to the study of spin polarized atoms and nuclei, and their applications. In most cases we used lasers to optically pump, and hence polarize, alkali-metal vapors.[1] Spin-exchange collisions were used to transfer the angular momentum to other systems.[2] Of particular interest was our continuing study of the polarization of noble gas nuclei, which are characterized by extremely long spin relaxation times of minutes to many days.

During this past research period we have demonstrated several applications of polarized noble gas nuclei that may have important implications for such diverse areas as non-destructive testing and medical diagnostics, as well as many areas in fundamental research. Of particular note is the use of spin-exchange polarized ^{3}He and ^{129}Xe for magnetic resonance imaging.[3] At present, our imaging work has focused on the lungs of small animals such as mice and guinea pigs. We believe, however, that our technique would also be useful for non-destructive testing. We have also continued our study of Xe that is polarized in the gaseous state, and subsequently frozen.[4] This novel technique for producing a highly polarized solid has received considerable attention in the NMR community.[5]

At the beginning of each of the following sections, we list the publications that have come out of our research. They are letter coded for easier reference within the text. We have also included as appendices three brief publications that appeared in letter format.

II. Magnetic Resonance Imaging

A. M. S. Albert, G. D. Cates, B. Driehuys, W. Happer, B. Saam, C. S. Springer Jr., and A. Wishnia, *Biological Magnetic Resonance Imaging Using Laser-Polarized* ^{129}Xe, Nature **370**, 199 (1994).

We have demonstrated that spin-exchange polarized noble gases can be used for a powerful new technique for magnetic resonance imaging (MRI).[3] Normally, MRI makes use of the protons which are prevalent in living organisms in both water and lipids. Imaging is possible because when the patient is placed in a large magnetic field, the protons become polarized due to the Boltzmann distribution:

$$P_p = \frac{N_\uparrow - N_\downarrow}{N_\uparrow + N_\downarrow} = \tanh\left(\frac{\mu_p B}{kT}\right) \approx \frac{\mu_p B}{kT}$$

At 1 Tesla and 310 K (body temperature) the resulting proton polarization

$$P_p = 3.29 \times 10^{-6}.$$

Typically, for a laser polarized noble gas:

$$P = 0.2 - 0.9.$$

This represents an increase in polarization of a factor of about 100,000! Not all of this gain is usable, however, because the density of spin in the gas is down by a factor of about

2700. The nuclear magnetic moments involved, are also somewhat smaller, particularly in the case of ^{129}Xe. The relevant nuclear magnetic moments are listed below.

- $\mu_p = 2.79\mu_N$
- $\mu_{He} = -2.13\mu_N$
- $\mu_{Xe} = -0.776\mu_N$

Taking all of this into account, and assuming a modest ^{3}He polarization $P_{He} = 0.2$, for a given fixed magnetic field we would have a gain in signal of 13. Assuming a modest Xe polarization $P_{Xe} = 0.2$, the gain is 1.7. Even so though, inert gas imaging has already demonstrated capabilities never before thought possible in MRI. Our images of lungs, for instance, are the best that have been obtained by any technique. In fact, for a variety of reasons, it is nearly impossible to image lungs using conventional MRI. Laser polarized inert gases represent a very different sort of contrast agent than has previously been used in MRI. Unlike water, they will only be present where they are introduced, such as in the lungs, if inhaled, or in the tissues fed by the blood. Also, they have very different MRI properties than do protons. The longitudinal and transverse relaxation times (T_1 and T_2) are different than protons, and in the case of Xe, large chemical shifts (100–300 ppm) exist depending on the environment in which the Xe resides.

Our imaging technique is a direct outgrowth of many of the experiments carried out in our group over the last ten years. The ^{3}He or ^{129}Xe is polarized in a sealed glass cell, containing 3–10 atmospheres of noble gas, about 50–75 Torr of nitrogen, and a few milligrams of alkali-metal, usually rubidium. The polarization of the sample as a function of time is characterized by the simple expression

$$P(t) = \frac{\gamma_{SE}}{\Gamma + \gamma_{SE}} \left(1 - e^{-(\Gamma + \gamma_{SE})t}\right) \langle P_{Rb} \rangle$$

where γ_{SE} is the spin exchange rate between the noble-gas and the Rb, and Γ is the relaxation rate of the noble gas spins due to everything other than the Rb. Here $\langle P_{Rb} \rangle$ is the average polarization of the Rb vapor. It is clear that high polarizations require maximizing γ_{SE}, and minimizing Γ. In this, we have the benefit of many years of research from which to draw. Once the noble gas is polarized, a small break-stem is broken, releasing the gas into a small surgical tube by which it is introduced to the animal. From this point on, the imaging is accomplished using techniques that are fairly standard in MRI.

MRI using laser polarized inert gases should also have applications in non-destructive testing. Some materials, such as ceramics, are difficult to image using techniques such as X-rays. One can imagine placing the part to be tested in a chamber, and subsequently flooding the chamber with a polarized inert gas. Any spaces within the ceramic, even those that are not readily visible, will fill with gas, and can then be imaged. Cracks or other defects should be readily visible as well.

II. Spin Interactions at Surfaces

B. B. Driehuys, G.D. Cates, W. Happer, H. Mabuchi, B. Saam, M. Albert, and A. Wishnia, *Spin Transfer Between Laser-Polarized* ^{129}Xe *Nuclei and Surface Protons*, Phys. Lett. A **184**, 88 (1993).

For many reasons, it is important to understand the interactions of spin polarized inert gases with various surfaces. For instance, our gaseous samples are typically contained in glass cells, and the relaxation that occurs at the walls of these cells often plays a critical role in determining the ultimate polarizations that are achievable, as well as the time scale over which the gases can be stored before they become depolarized.

One intriguing possibility is that various types of atoms and nuclei at surfaces may be polarized by coming into contact with nuclear polarized inert gas. We have demonstrated such a "cross-polarization" technique by polarizing the protons in a silicone coating through contact with polarized ^{129}Xe.[6] The silicone coating, Surfasil, is a coating that we frequently use to suppress wall relaxation in cells containing ^{129}Xe. Our experiment utilized a sealed glass cell, whose interior surface was coated with Surfasil. The cell was filled with 400 Torr of isotopically enriched Xe (72.9% ^{129}Xe), several milligrams of Rb, and 75 Torr of N_2. After polarizing the ^{129}Xe, the cell is cooled in liquid nitrogen for about 10 seconds in a field of about 40 Gauss, and subsequently kept in the earth's field (of about 0.5 Gauss) for about 3 seconds. The cell is then quickly placed in a pulsed NMR aparatus where the proton signal is observed.

This work has several interesting implications. First, in conjunction with the imaging experiments described in the previous section, it may be possible to produce images of specific nuclei in surfaces! This could be of great use for non-destructive testing of coatings, particularly when the coatings reside on inaccessible surfaces. This work also led us, after considerable work, to discover that the mechanism that causes spin relaxation on Surfasil is in fact cross-relaxation to the protons in the methyl groups of the Surfasil. This important discovery, which will probably lead to greatly improved surface coatings, occurred during the subsequent research period and will be described in the next technical report.

Frozen Polarized Xenon

C. M. Gatzke, G. D. Cates, B. Driehuys, D. Fox, W. Happer, and B. Saam, *Extraordinarily slow nuclear spin relaxation in frozen laser-polarized* ^{129}Xe, Phys. Rev. Lett. **70**, 690 (1993).

In 1990, we published a letter in which we showed that Xe, after being polarized by spin exchange, could be frozen with little or no loss of polarization.[7] During this past research period, we have continued to investigate frozen Xe, and have determined the predominant spin-relaxation mechanisms over a wide range of temperatures.

For temperatures of roughly 20–120 K, spin relaxation occurs because of a Raman scattering of phonons due to a spin rotation interaction of the form

$$\mathcal{H}_{\mathrm{sr}} = \gamma_I \mathbf{I} \cdot \mathbf{N}$$

where $\mathbf{I}$ is the spin of a ^{129}Xe nucleus, $\mathbf{N}$ is the relative angular momentum of a pair of neighboring atoms, and γ_I characterizes the strength of the interaction. At temperatures of 77 K, this interaction causes relaxation with time scales of several hours. At lower temperatures, as the hot phonon spectrum freezes out, the relaxation times due to $\mathcal{H}_{\mathrm{sr}}$ are expected to be very long, longer in fact than we observed.

At temperatures below about 20 K, it is our belief that relaxation occurs largely because of cross-relaxation to ^{131}Xe. We convinced ourselves of this through several experiments. For instance, we demonstrated that simply lowering the applied magnetic field to zero (leaving only the earth's field) had the effect of transferring polarization from the ^{129}Xe nuclei to the ^{131}Xe nuclei. While this was very suggestive, it still remained to demonstrate that at high fields cross-polarization was still taking place. Toward this end, we studied samples in which we varied the fraction of ^{131}Xe. We found a striking dependence indicating that the less ^{131}Xe in the sample, the less relaxation we observed. At high fields, we believe that the relaxation occurs largely at the surfaces of the Xe crystallites. At such locations their are large electric field gradients which can cause large quadrupolar splittings, which help facilitate the relaxation.

Spin exchange and spin relaxation in the bulk phase

D. N. R. Newbury, A. S. Barton, G. D. Cates, W. Happer, and H. Middleton, *Gaseous ^{3}He–^{3}He Magnetic Dipolar Spin Relaxation*, Phys. Rev. A **48**, 4411 (1993).

E. N. R. Newbury, A. S. Barton, P. Bogorad, G. D. Cates, M. Gatzke, H. Mabuchi, and B. Saam, *Polarization dependent frequency shifts from* Rb–^{3}He *collisions*, Phys. Rev. A, **48**, 558 (1993).

F. A. S. Barton, N. R. Newbury, G. D. Cates, B. Driehuys, H. Middleton, and B. Saam, *Self-calibrating measurement of polarization-dependent frequency shifts from* Rb–^{3}He *collisions*, Phys. Rev. A **49**, 2766 (1994).

While the spin interactions of alkali-metal atoms and noble-gas atoms are certainly best understood in the gaseous phase, there still remain various important points of interest that we continue to investigate.

Perhaps the most important result of this research period in this area is described in publication "D" above. As mentioned earlier, a critical factor in both the polarization and storage of polarized noble gases is the spin-relaxation time of the gas. In the case of ^{3}He, it is possible to achieve sufficiently non-relaxing walls that the predominant mechanism is spin relaxation in ^{3}He–^{3}He collisions due to dipolar interactions. We have studied this relaxation both theoretically, and experimentally, and found that the relaxation time for a density of 10 Amagats is 74.4 hours, and varies inversely with density. This important number has become a benchmark that we seek to obtain whenever fabricating ^{3}He cells. In fact, we have found that by using extreme care, we can often come quite close to this "bulk limited" result.

Two more studies, described in publications **E** and **F** explore the phenomena of frequency shifts caused in the electron paramagnetic resonance (EPR) spectrum of Rb atoms due to being in the presence of nuclear polarized ^{3}He. Since the shift in the EPR frequency depends on the Rb polarization, the study of such frequency shifts has great promise as a new form of polarimetry. Also, the atomic parameter that characterizes these frequency shifts carries important information concerning the spin-exchange interaction, which is interesting in and of itself.

Collaborations with Outside groups

G. P. L. Anthony *et al.*, *Determination of the neutron spin structure function*, Phys. Rev. Lett. **71**, 959 (1993).

H. A. S. Barton, P. Bogorad, G. D. Cates, H. Mabuchi, H. Middleton, N. R. Newbury, R. Holmes, J. McCracken, P. A. Souder, J. Xu, and D. Tupa, *Highly polarized muonic He produced by collisions with laser optically pumped Rb*, Phys. Rev. Lett. **70**, 758 (1993).

There are several activities at Princeton, supported by separate NSF and DOE grants, that utilize optical pumping and spin-exchange polarized noble gases for the investigation of various subatomic phenomena. These efforts often have a productive symbiotic relationship to AFOSR supported activities since they share many of the same technical concerns. This was particularly true of an experiment at the Stanford Linear Accelerator Center (SLAC) to study the origin of spin in the neutron. The experiment, which is described in publication "G", utilized a polarized ^{3}He target. Much of the work that made this experiment possible translated directly into making our new MRI work possible. Also, much of the laser technology developed for the experiment described in publication "H" was also useful. Several people who were supported in part by the AFOSR participated sufficiently in these efforts to be included as coauthors on the resulting publications.

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VIII. Personnel

The people listed below received full or partial support from the grant during some portion of the period covered by this report.

Faculty
Dr. William Happer, Professor of Physics
Dr. Gordon Cates, Associate Professor of Physics

Post Doctoral Research Associates
Dr. Hunter Middleton

Graduate students

Note, the name marked with a ⋆ received a Ph.D. from work supported by this grant. Also, the names marked with a • are soon to receive Ph.D.'s from work supported by this grant.

⋆ Dr. Michael Gatzke (received Ph.D. in August 1992, now at the University of Virginia)
• Mr. Brian Saam
• Mr. Bastian Driehuys
   Dr. Nathan Newbury (received Ph.D. in August 1992, now at JILA)
   Ms. Amy Barton

Undergraduates who worked on Princeton senior theses
Mr. David Fox, (now at Harvard, Apker Award Finalist)
Mr. Jordan Katine (now at Harvard)
Mr. Hideo Mabuchi (now at Cal Tech)